

Development of an Atmospheric Mercury Modeling System for the Great Lakes Region

Progress Report for the Quarterly Period Ending September 30, 2002

Wisconsin Department of Natural Resources
Mercury Analysis Team

The Mercury Analysis Team, part of the Wisconsin Department of Natural Resources' (DNR) Air Management Program, is responsible for developing an atmospheric mercury modeling system for Wisconsin and the Great Lakes region. Partial funding for this effort comes from a grant awarded by USEPA in October 2001. The team identified seven major areas of work and the lead staff for each as follows:

- Atmospheric Chemistry Modeling, Mike Majewski – WDNR
- Meteorological Modeling, Wusheng Ji – WDNR
- Regional Emission Modeling, Gwendolyn Judson – WDNR
- Mercury Inventory Development, Orlando Cabrera-Rivera & Grant Hetherington – WDNR
- Data Analyses, William Adamski, Grace Liu & Sanobar Durrani – WDNR
- Mercury Monitoring, Mark Allen – WDNR
- Computer Resources, Mike Majewski – WDNR

The Team meets on a regular basis and is the coordinating body for this project providing staff and other interested parties the opportunity to contribute feedback and ideas. Meeting minutes can be found at www.dnr.state.wi.us/org/aw/air/staff/hganalysisteam/index.htm. This document records our progress in each major area for the quarter ending September 30, 2002.

Atmospheric Chemistry Modeling

Modeling activity has centered on testing several different versions of the REMSAD model, version 6.40 and version 7.02.

According to ICF Consulting, version 6.40 includes updates/corrections to the sulfate formation and wet deposition algorithms. Also, limited testing for mercury indicates improvements for Hg deposition as well. Version 7.02 includes new treatment of secondary organic aerosols.

Version 7.02 represents the third version of the REMSAD model we have run. We have run all three versions of the model with data for the month of July 1996. This is the original data that ICF Consulting used in the REMSAD mercury modeling for LADCO.

Some maximum values (units = g/km²) are:

- Monthly total dry deposition of THG...43.7(v6.20)...43.3(v6.40)...43.0(v7.02)
- Monthly total wet deposition of THG...9.4(v6.20)...16.3(v6.40)...16.3(v7.02)
- Monthly total wet + dry deposition of THG...47.1(v6.20)...49.0(v6.40)...48.6(v7.02)

The location of the total “wet + dry deposition of THG” maximum has not changed for any of the versions. This location is in northeast Indiana.

The greatest change is the increase in “total wet deposition of THG” between versions v6.20 and v6.40.

In Wisconsin there are some increases in the “total wet + dry deposition of THG”. These are in the west central part of the state and appear to be on the order of 1 g/km². Generally, the model changes have greater impacts at locations in the southern Plains, southeastern states, and along the East Coast

Meteorological Modeling

In this Quarter we first have migrated our MM5 model system including all initialization files from a Deck Alpha workstation to a Red Hat Linux with six processors, which enables us to run our MM5 model with a superior simulation domain. Secondly, since the ongoing studies have demonstrated that the daily rainfall is directly responsible for mercury’s wet deposition and the model rainfall forecast is also very sensitive to the specification of the model cumulus scheme, therefore, we had conducted a series of sensitivity tests trying to determine the best cumulus scheme for our modeling application.

With an access to a more powerful workstation, our sensitivity studies were conducted with the LADCO/Midwest Regional Planning Organization (RPO) 36km grid as the coarse grid and an imbedded 12km grid as the fine grid, as shown by Fig. 1. The coarse, RPO national scale 36km grid had 165X129 grid points and covered the most part of North America. The inner 12km grid had 169X169 grid points with the domain centered over Wisconsin and covered in an area from Texas at southwest corner, South Carolina at southeast, Saskatchewan at northwest and Quebec at northeast. Both grids engaged a properly selected vertical mesh size in order for the model to resolve the atmospheric thunderstorms more accurately. The model has 41 vertical layers with 116 m vertical spacing in the lowest level, gradually stretching upward to about 688 m by the top level near 17km with over 20 layers reserved in the middle for rainfall system evolution.

The MM5 version3.5 had a total of eight different cumulus parameterizations, of which four are suitable for our model application of grid sizes of 36km and 12km, which are:

- 1) Grell parameterization, based on the rate of destabilization and simple single-cloud scheme;
- 2) Fritsch-Chappell parameterization, base on relaxation to a profile due to updraft, downdraft and subsidence region properties;
- 3) Kain-Fritsch parameterization, similar to Fritsch-Chappell, but used a cloud-mixing scheme to remove all available buoyant energy in the relaxation time;
- 4) and finally the Kain-Fritsch 2 parameterization, which is a new version of Kain-Fritsch that includes shallow convection.

Unfortunately, our sensitivity test runs had only finished with the first three parameterizations at this time. The last one, Kain-Fritsch 2 test run was interrupted in the middle and its run should be finalized shortly. Therefore, this report will only cover the first three results. The atmospheric data used for initializing the MM5 as well as in the lateral boundary and 4DDA nudging schemes were analyzed with the NMC 2.5 mandatory level analysis and the conventional surface and upper air datasets obtained from the archives of the National Center for Atmospheric Research. The standard NWS rawinsondes at 0000UTC and 1200 UTC were used in the nudging procedures. Due to dominated synoptic features of a long last stationary front and the precipitation during the episode, the explicit cloud and precipitation options were all turned into active for this modeling simulation. The explicit moisture schemes of simple ice, high-resolution

Blackadar PBL scheme and cloud radiation scheme were employed for the simulation. The model run starts at 1200UTC on June 21, 1998, ends at 1200UTC on June 30, 1998 with a restart at 1200UTC on June 25, 1998, and takes about 10 days to simulate the episode with a Red Hat Linux workstation.

Based on the rainfall pattern and the daily rainfall amount during the episode, and the comparisons with the NOAA daily weather precipitation maps, it seems that the Grell scheme had produced the most accurate rainfall estimate for the seven-day episode between June 23-29, 1998. The Kain-Chappell scheme produced the worst, and the Fritsch-Fritsch is in the middle. Fig. 2 shows the heaviest rainfall day for the episode, June 27, 1998. It indicates that the majority part of Minnesota/Wisconsin had received at least one inch of rainfall by 7:00am EST on that day, of which a small portion received at least two inches with maximum of 2.54 over St. Paul/Minneapolis. Fig. 3 is the 24 hours of rainfall forecast produced by the MM5 model with the Fritsch-Chappell cumulus parameterization. Even through the Fritsch-Chappell scheme had reproduced a considerable amount of rainfall over the Minnesota/Wisconsin region, the biggest dilemma lies over the Arkansas/Tennessee region, where the model showed heavy precipitation with maximum of 3.54 inches over the region while the observation showed nothing. Fig. 4 shows the same day precipitation generated by the Grell scheme, which indicates that the Grell scheme had successfully reproduced the rainfall pattern for the Minnesota/Wisconsin region in the 12km grid, and it also had generated a reasonable rainfall distribution outside the 12km grid. However, the setback with the Grell scheme is that it seems to have a tendency of overestimate the rainfall amount, such as show by Fig. 5. The map shows that there was a rainfall center over South Dakota with maximum of 5.74 inches, which could not be confirmed neither by the observation, nor by the any other test run, such as Kain-Fritsch shown by Fig. 6. Although the Kain-Fritsch scheme had displayed a more reasonable result for this heavy rainfall day, it still has difficulty with the light rainfall day. Fig. 7 is the observation for the lightest rainfall day during the episode, June 26, 1998, which shows there is only trace amount of rainfall for the 12km grid region. However, the Kain-Fritsch scheme still produced certain amount of heavy rainfall for those areas with no precipitation at all, as shown by Fig. 8. Whereas, the Grell scheme shown by Fig. 9 had displayed a more sensible rainfall pattern.

Based on our sensitivity tests so far, it seems that the Grell scheme is the most accurate one among those three schemes for rainfall forecasting, and the Kain-Chappell is the worst. Since we are planning to resume our test run with Kain-Fritsch 2 scheme soon, it would be very interesting to see what its results could be. In the near future it would be also very nice to use a different initialization dataset like Eta model output, or use a different moisture scheme like mix-phase to further study the model's capability of predicting the daily rainfall, but we may not have time or resources to test them all.

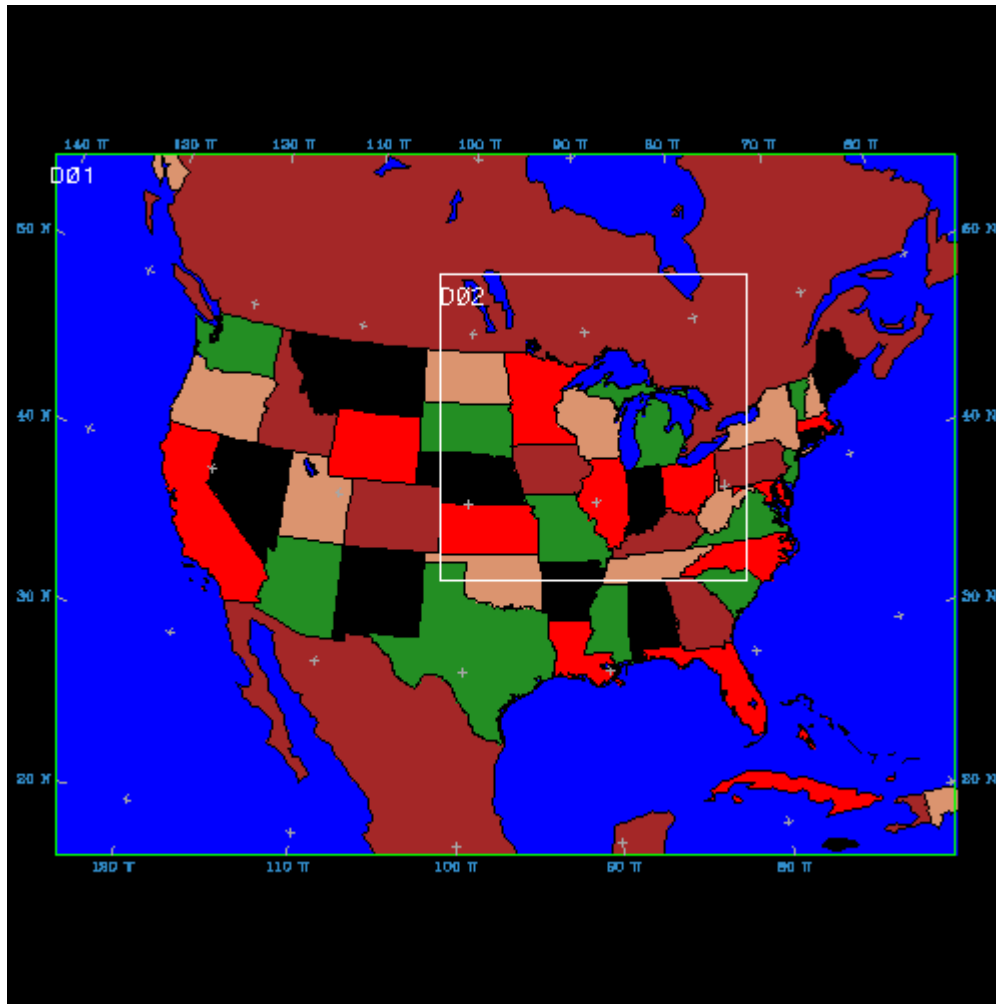


Fig. 1. The MM5 modeling domains for June 1998 episode showing the RPO national 36km grid (D01) and the inner 12km grid (D02) with the grid centering over Wisconsin.

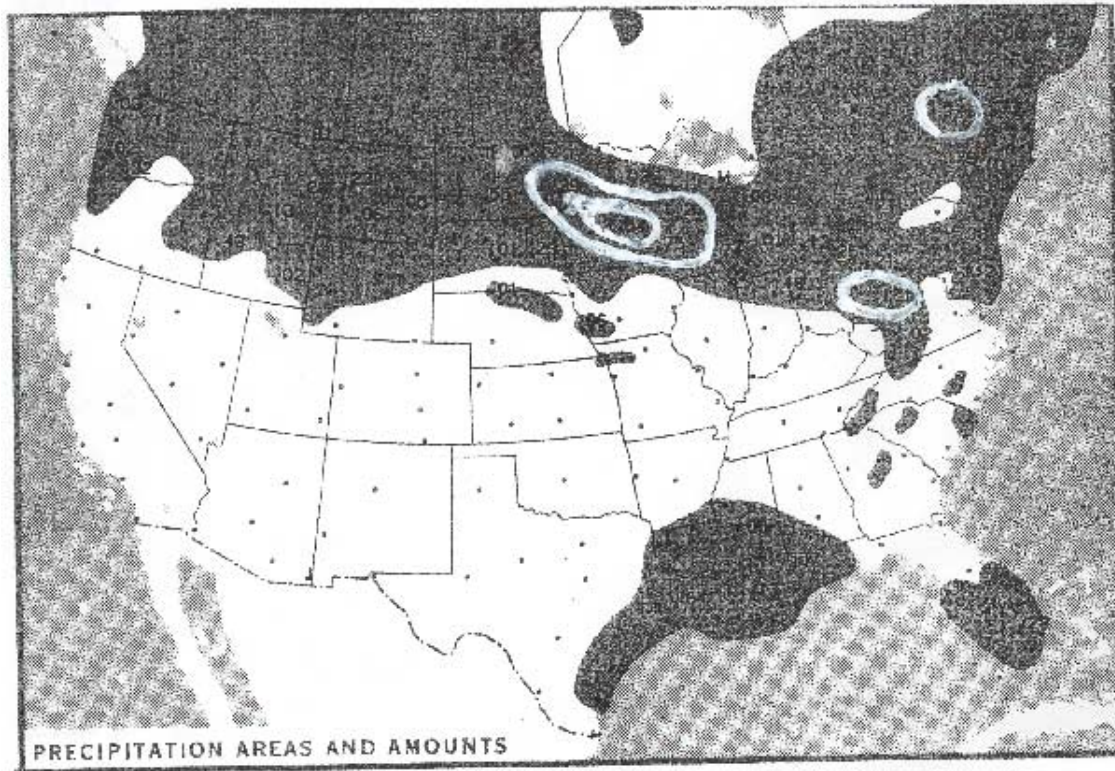


Fig. 2 NOAA daily weather maps of precipitation for June 27, 1998. Shaded areas show at least traced amount of precipitation during the previous 24 hours ending at 7:00am, EST. The white contour lines in the shaded areas show the precipitation amount with the one-inch interval. The maximum amount of rainfall is 2.54 inches in the center of the two-inch contour line over St. Paul/Minneapolis.

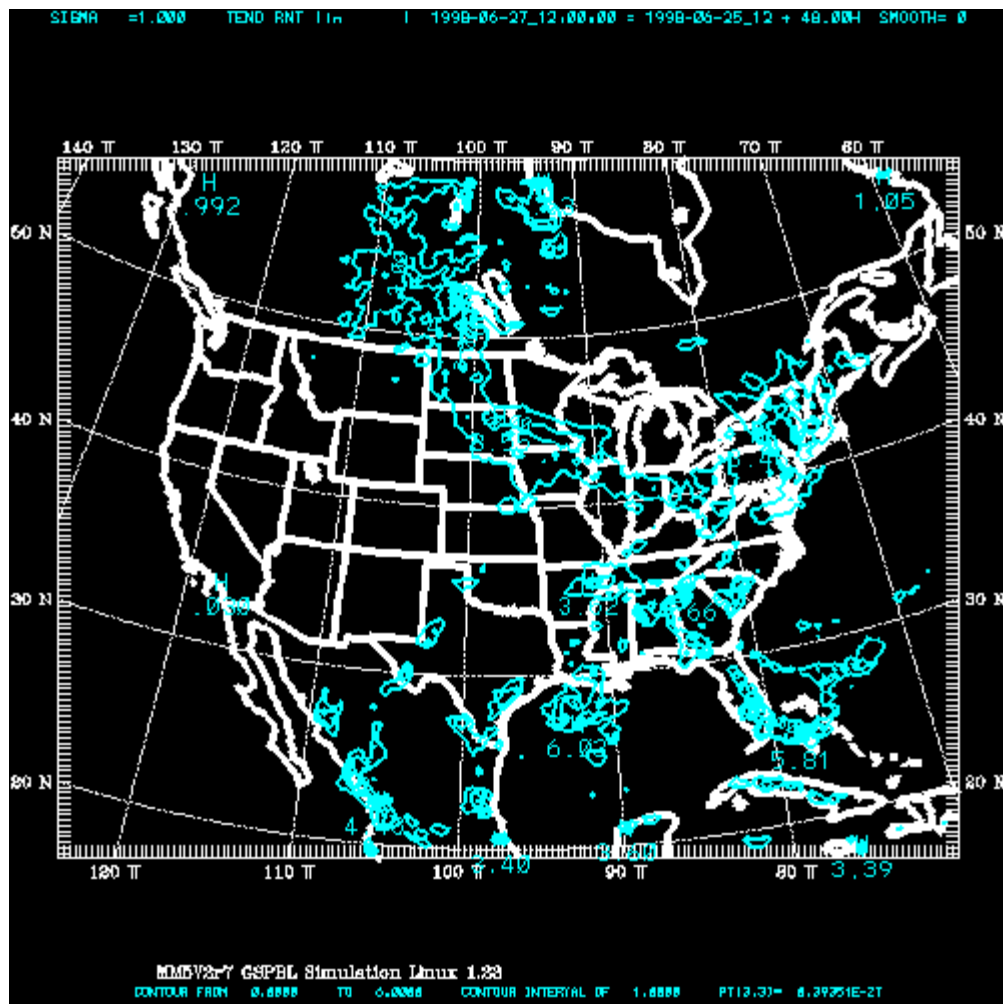


Fig. 3. MM5 model with Fritsch-Chappell parameterization generated daily precipitation in the unit of inch during the front-induced rainfall of June 27, 1998 at 7:00am EST for the 36km grid.

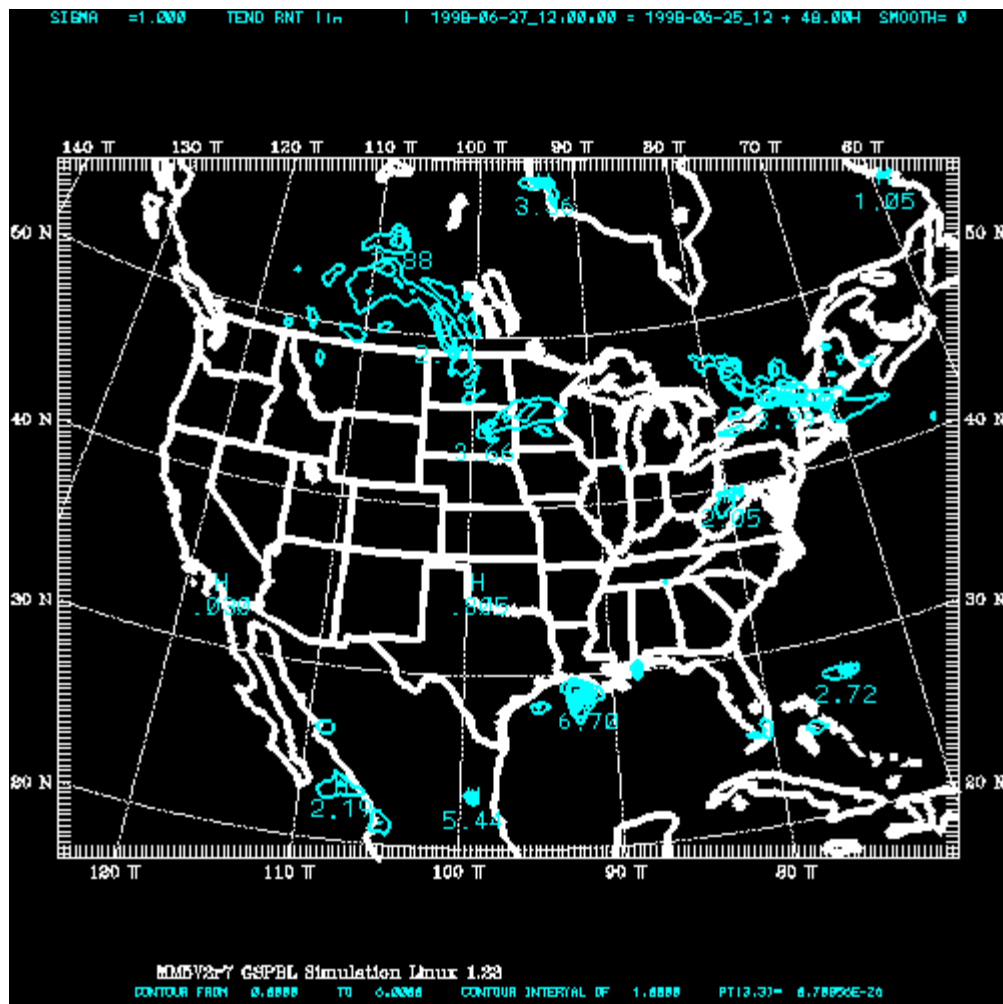


Fig. 4. MM5 model with Grell parameterization generated daily precipitation in the unit of inch during the front-induced rainfall of June 27, 1998 at 7:00am EST for the 36km grid.

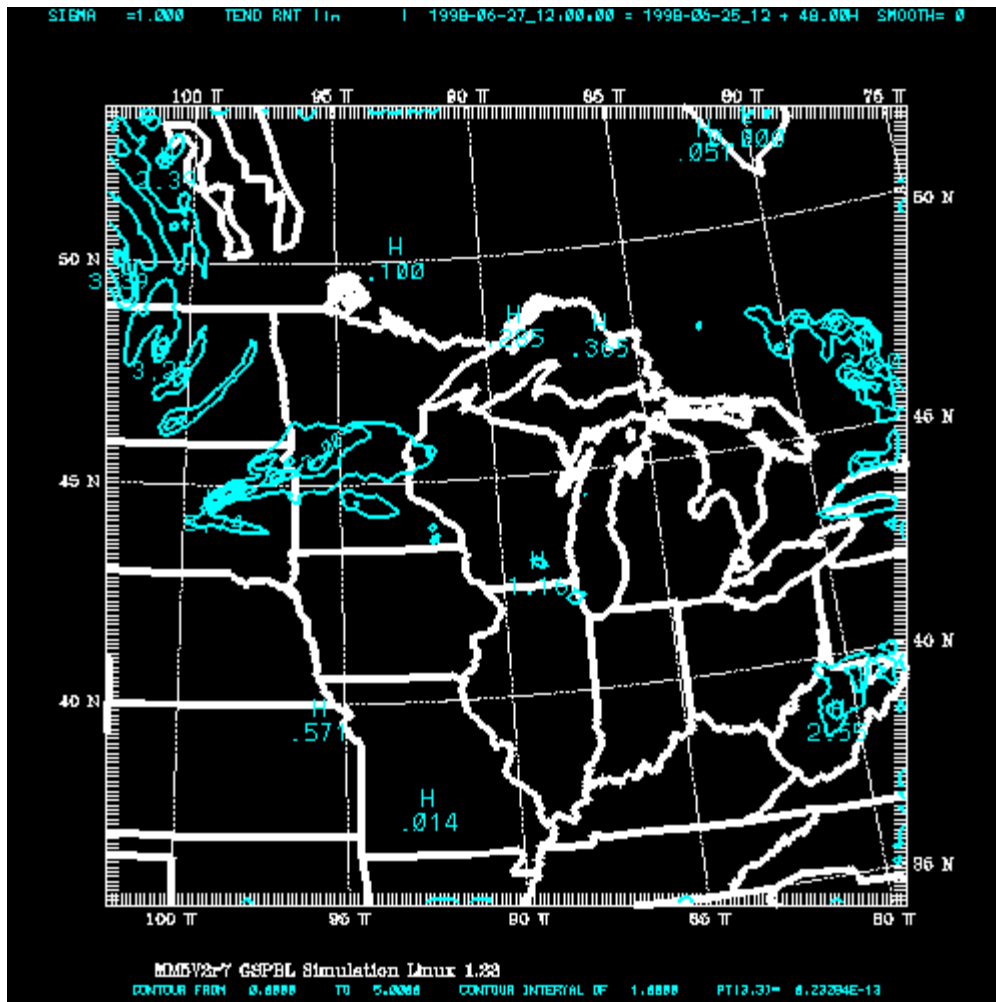


Fig. 5. MM5 model with Grell parameterization generated daily precipitation in the unit of inch during the front-induced rainfall of June 27, 1998 at 7:00am EST for the 12km grid.

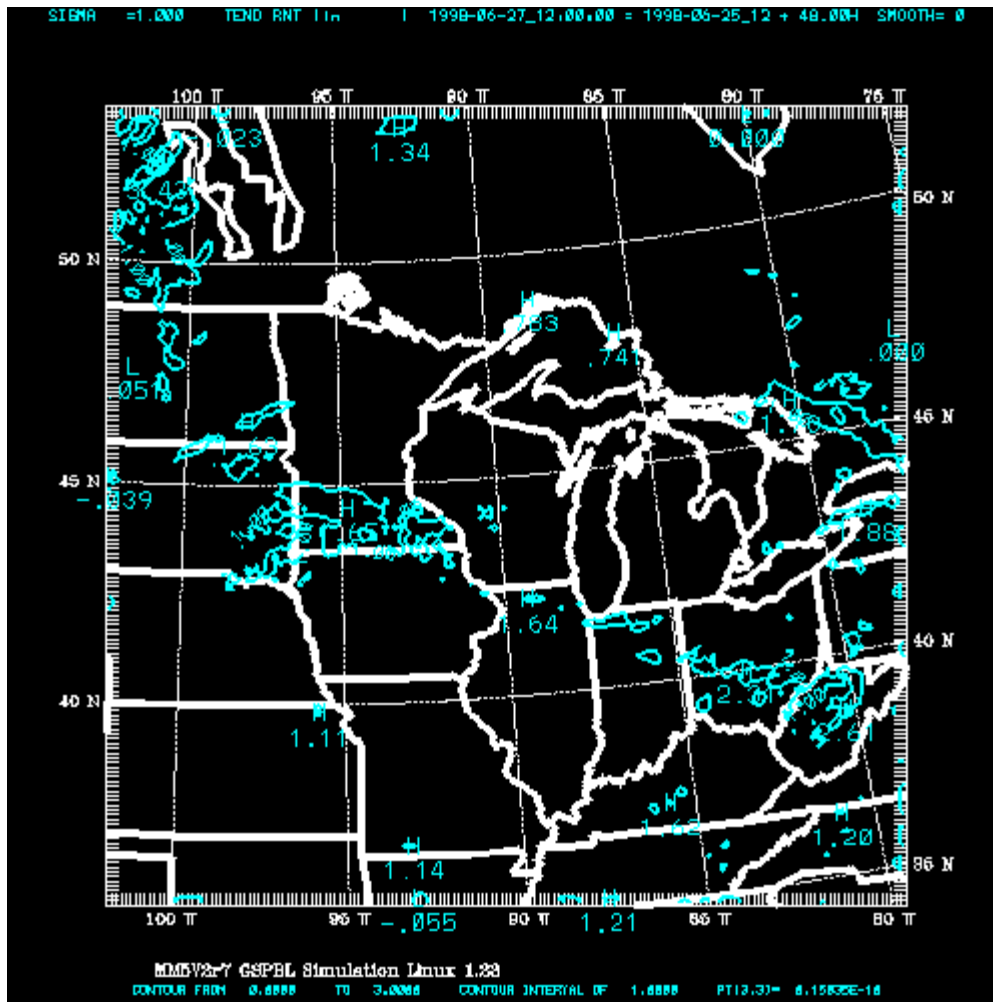


Fig. 6. MM5 model with Kain-Fritsch parameterization generated daily precipitation in the unit of inch during the front-induced rainfall of June 27, 1998 at 7:00am EST for the 12km grid.

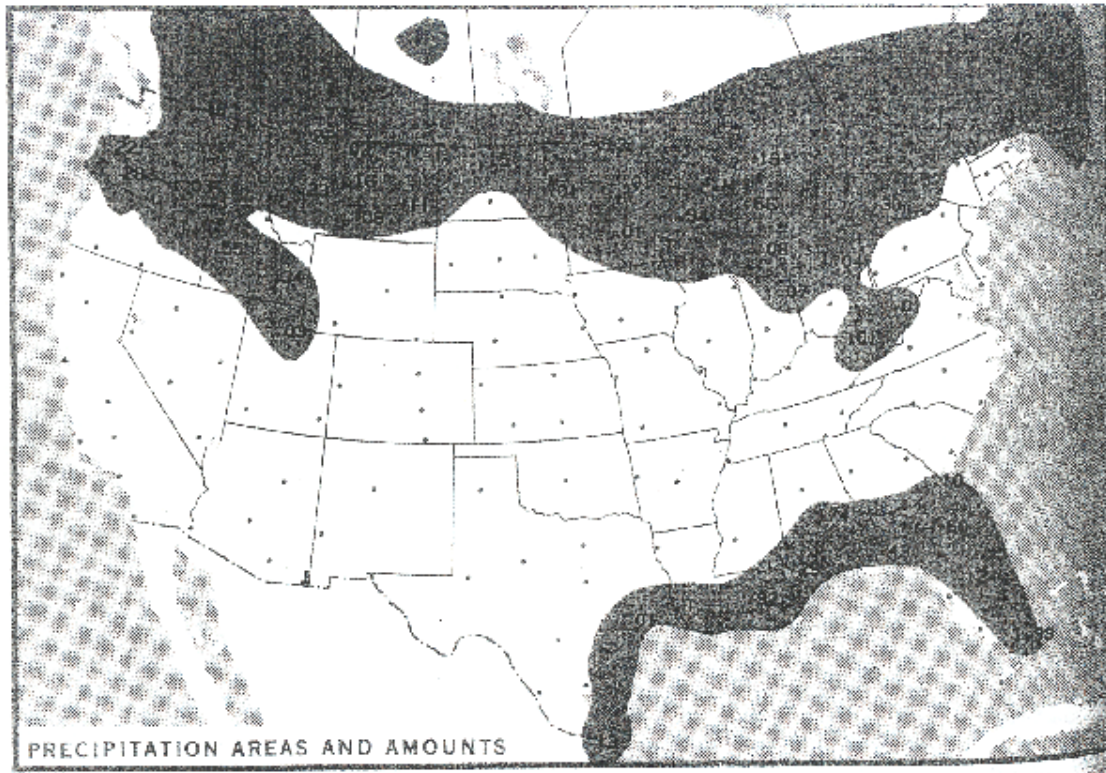


Fig. 7. NOAA daily weather maps of precipitation for June 26, 1998. Shaded areas show at least traced amount of precipitation during the previous 24 hours ending at 7:00am, EST. The majority of the precipitation region only received traced amount of rainfall during that day.

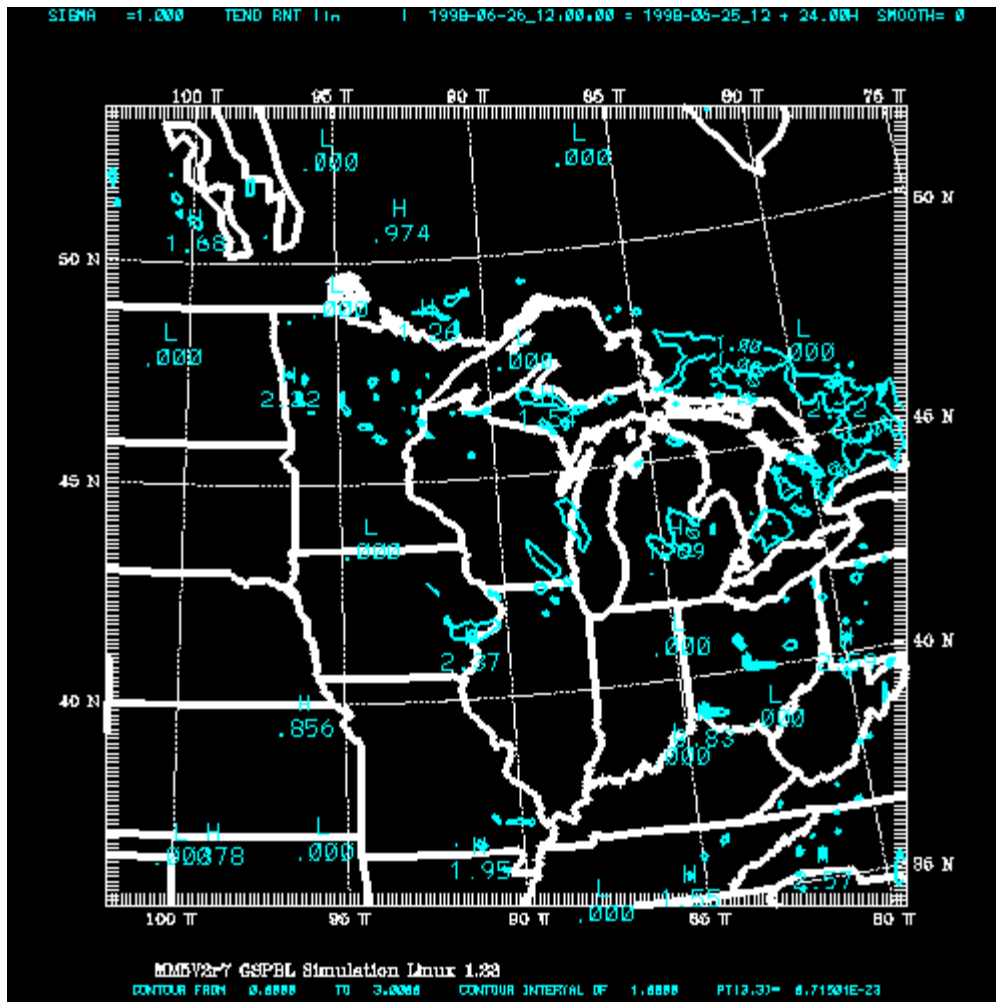


Fig. 8. MM5 model with Kain-Fritsch parameterization generated daily precipitation in the unit of inch for June 26, 1998 at 7:00am EST with the 12km grid.

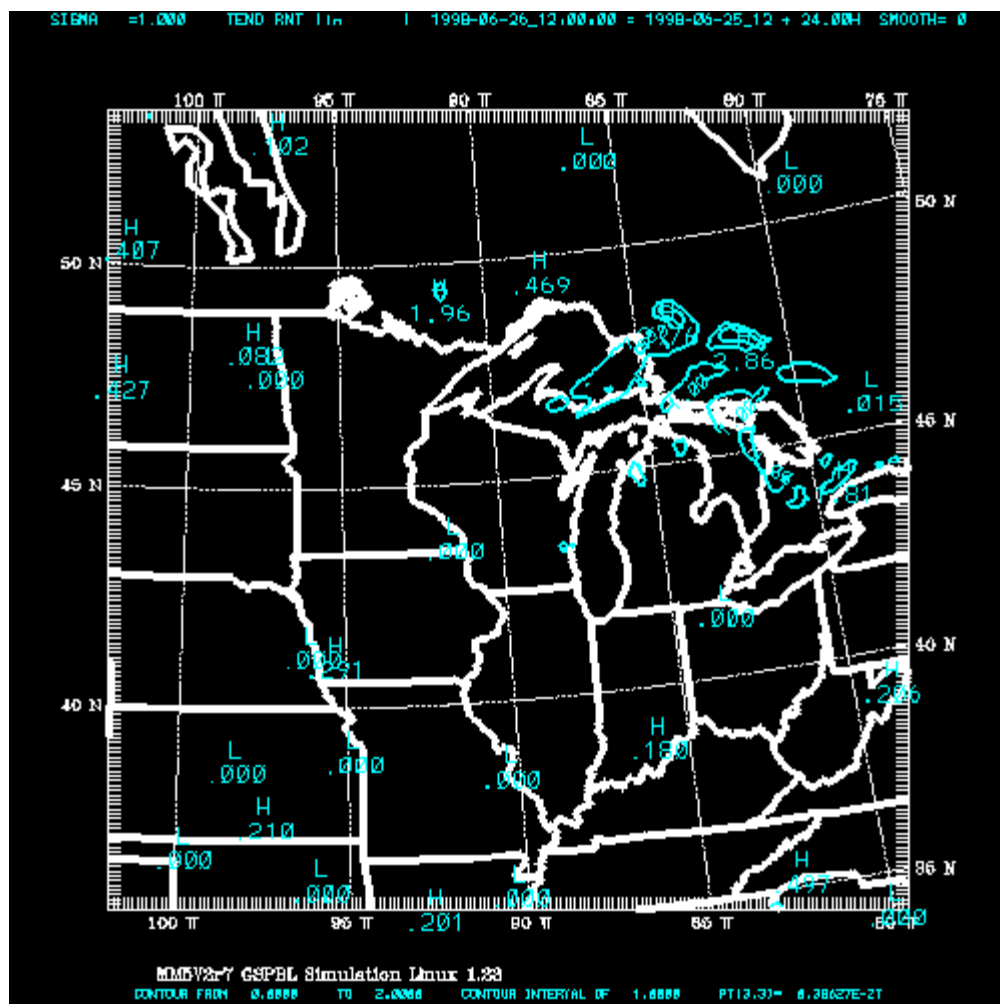


Fig. 9. MM5 model with Grell parameterization generated daily precipitation in the unit of inch for June 26, 1998 at 7:00am EST with the 12km grid.

Regional Emissions Modeling

The mercury species needed for chemistry modeling have been incorporated into the EMS-2001 databases and model code. Version 2.0 of the 1999 NEI for hazardous air pollutants has been downloaded from USEPA and is being used to test the new processors.

Based on the quality of the data in the NEI, new reports to enhance quality assurance will be developed for EMS-2001 in the next 6 months.

All files and directories needed to run the national grid being used by LADCO for their ozone and particulate runs have been added.

Inventory Development

Several refinements to the 1999 Wisconsin mercury emissions inventory were completed this quarter.

- Identified ninety-three stationary combustion sources that did not report mercury to the emissions inventory. The emissions from these sources were estimated using EPA's Factor Information Retrieval data system (FIRE) version 6.23 emission factors. The estimates resulted in emissions below the reporting threshold under chap. NR 438. Emissions from these sources totaled 19 lb.
- Calculated mercury emissions from fluorescent lamp breakage and recycling. Emissions for all counties from lamp breakage were calculated. Emissions from lamp recycling were calculated for counties with fluorescent lamp recycling processing facilities. Sites being used exclusively for drop-off and collection of fluorescent lamps were excluded. Emissions from these sources totaled 31 lb.
- Calculated mercury emissions from forest fires. These emissions were estimated using an emission factor of 71.3 ng/g biomass. This factor was obtained from a National Center for Atmospheric Research (NCAR) research paper. The resulting emissions for Wisconsin amounted to 8.6 lb.
- Corrected mercury emissions for twelve paper facilities. Reported mercury emissions were corrected based on the Mercury Advisory Group emission estimates for industrial boilers. The revised estimates resulted in equal or lower emissions from those reported.
- Speciation profiles have been assigned to all SCCs expected in the mercury inventory. The profiles were compiled from previous modeling reports. For each category, the most recent available data was used with consideration given to the documentation available of its source. Research in this area will continue.

In order to assess the magnitude of mercury emissions from poorly quantified potentially significant types of sources, measurements of ambient air mercury concentrations have been performed near lamp recycling facilities and diesel trucks. In the future, further measurements will be made at other sources including crematoriums and limekilns.

No final version 2 of the 1999 National Emissions Inventory (NEI) was released in June. EPA has decided not to finalized version 2 of the 1999 NEI. Instead, EPA plans on releasing a preliminary version 3 of the 1999 NEI in the beginning of October. In anticipation of this release, the QA plan has been revised and refined. A list of all source classification codes (SCCs) used to report mercury emissions by the Great Lake States or having mercury emission factors associated with them has been created as a screening QA tool.

Data Analyses

There are no updates for this topic at this time.

Mercury Monitoring

The Wisconsin DNR's Air Program continued an active program for mercury monitoring in the third calendar quarter of 2002. Deposition monitoring for mercury continued at five sites in Wisconsin. Ambient mercury monitoring was conducted at ground stations and from an aircraft. A summary of the monitoring projects follows.

Deposition Monitoring

Wisconsin has five monitoring stations as part of the National Mercury Deposition Network (MDN) operated by the National Atmospheric Deposition Program (NADP). The sites are located at Brule River, Trout Lake, Lake Du Bay, Devils Lake, and Lake Geneva. Four of these sites collect weekly wet deposition samples. A fifth site, at Devils Lake, is operated as an event site where the sample is removed from the collector after each rainfall event. Information about the mercury deposition program as well as historical data for the Wisconsin monitoring stations can be found at the National Atmospheric Deposition Programs web site (<http://nadp.sws.uiuc.edu/>).

Ambient Monitoring

Mercury surveys continued using the portable LUMEX analyzer. This real time analyzer uses spectrophotometric principles to measure mercury in the air. The LUMEX has both a quick response and high sensitivity with a detection limit of 2 ng/m³ of air. The analyzer is subject to periodic baseline drift that limits its usefulness for long-term unattended operations. LUMEX surveys in the third quarter focused on fluorescent light recyclers. Results indicate that mercury emissions are variable. The most significant variable affecting mercury emissions is the on-site crushing of the light bulbs.

A report on Monitoring near the Mercury Waste Solutions facility in Union Grove was released and is available on the DNR's web site at

[*http://www.dnr.state.wi.us/org/aw/air/monitor/hgmonuniongrove.pdf*](http://www.dnr.state.wi.us/org/aw/air/monitor/hgmonuniongrove.pdf)

The report covers a short-term air monitoring project at the facility from April 4, 2002 to May 16, 2002.

The Mercury Analysis Trailer (MAT) shared with Michigan and Minnesota was not available to the WDNR during the third calendar quarter. The WDNR was able to use some of the equipment from the MAT. This equipment included one TEKRAN 2537a analyzer and supporting meteorological equipment. The TEKRAN analyzer was used for a study at a monitoring trailer near the Vulcan chemical plant in Port Edwards. Vulcan Chemical is the only chlor-alkyl plant in Wisconsin using mercury filled cells.

Aircraft Monitoring

August 30 was the start of a monitoring project using gold traps for long duration mercury sampling from an aircraft. The gold traps are commercially prepared glass tubes filled with gold-coated sand. The tubes will trap mercury from air drawn through the tubes. At the analysis laboratory, the mercury is thermally desorbed from the gold and the mercury measured using an atomic fluorescence analyzer. The analysis follows the protocol in USEPA Method IO-5.

Samples will be collected on periodic (approximately 1-in 12 days) aircraft flights from August 2002 until February 2003. The flights will measure mercury in the air above Lake Superior.

Computer Resources

The Lake Michigan Air Directors Consortium with funding from Wisconsin DNR and Illinois EPA have contracted with Environ to implement mercury chemistry in CAMx. The work is scheduled to be complete in 6-7 months. We expect to use this new version of the model when it becomes available.